

Instability of charge ordered states in doped antiferromagnets

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We analyze the induced interactions between localized holes in *weakly-doped* Heisenberg antiferromagnets due to the modification of the quantum zero point spin wave energy; *i.e.* the analogue of the Casimir effect. We show that this interaction is *uniformly attractive* and falls off as r^{-2d+1} in d dimensions. For “stripes”, *i.e.* parallel $(d-1)$ -dimensional hypersurfaces of localized holes, the interaction energy per unit hyperarea is attractive and falls, generically, like r^{-d} . We argue that, in the absence of a long-range Coulomb repulsion between holes, this interaction leads to an instability of *any* charge-ordered state in the dilute doping limit.

It is still not clear what happens when a dilute concentration of holes is introduced into a quantum Heisenberg antiferromagnet (AF). This is one of the central issues in the theory of correlated electronic systems, especially as it relates [1] to the high temperature cuprate superconductors and related oxides. One class of proposals [2–4] holds that the result is a spatially inhomogeneous “charge ordered” ground state. Unfortunately, numerical analysis of the stability of such states is often inconclusive because the typical energy differences between states are small, and the Goldstone modes (spin-waves) produce finite size effects which decrease slowly with system size.

The goal of this paper is to investigate the thermodynamic stability of charge-ordered states in short-range AF spin models in the dilute doping limit. We calculate the induced interaction between well-separated clusters of holes due to their modification of the spin wave spectrum, and find that it is *uniformly attractive*. Specifically, the asymptotic long distance ($r \rightarrow \infty$) interaction between two hole clusters (see Fig. 1) is of the form $\mathcal{E} \sim -JSr^{-2d+1}$ for a d -dimensional, spin- S Heisenberg quantum AF with exchange coupling J (see Eq. 2). For extended clusters of holes, under generic circumstances, the dependence on separation (but not the absolute magnitude) of the interaction energy can be reliably estimated by summing the pairwise hole-hole interaction over all pairs; for example, the interaction per unit hyperarea between parallel walls of localized holes (*i.e.* codimension 1 hypersurfaces which, with the case of $d=2$ in mind, we refer to as “stripes”) falls with their separation as $\mathcal{E} \sim -JSr^{-d}$.

As a consequence of this attraction, in the absence of a long-range Coulomb repulsion between holes, all charge-ordered states with sufficiently small hole concentration are unstable to phase separation, although it is possible that there exist non-vanishing hole densities for which charge ordered states are stable [3,4]. Remarkably, we find that the correct asymptotic form of the induced interactions cannot be obtained in any finite order of naïve perturbation theory, because of the singular effect of a marginally bound (zero-energy) spin-wave state associated with the fact that doped holes actually change the Hilbert space, by changing the number of spins. How-

ever, a simple modified perturbation theory can be constructed which qualitatively reproduces the exact spin-wave results in all cases we have tested.

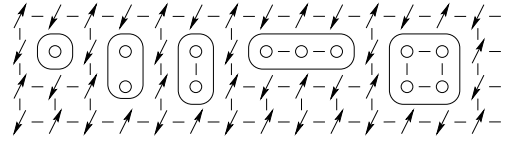


FIG. 1. Clusters of localized holes are modeled by removing the bonds connecting the spins to the rest of the system. The additional bonds inside the disconnected areas do not contribute to the interaction between hole clusters.

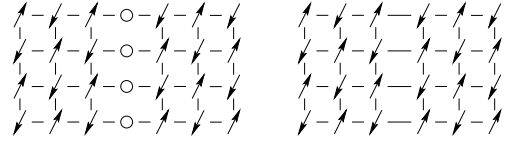


FIG. 2. A stripe of holes forming an anti-phase domain wall is modeled as a line of weak bonds in a perfect AF.

The Model: The number of spin degrees of freedom changes with doping and, therefore, the Hilbert spaces appropriate to the doped and undoped AF are different. Therefore, for mathematical convenience it is preferable to use a model with a spin S operator on each site, and to treat any system with localized holes as a limiting case in which the coupling between a set of “impurity” sites and its neighbors goes to zero. For static holes, this is all there is to the model. However, so long as the holes are *localized*, either by an impurity potential or by a self-consistent field (as in Hartree-Fock solutions [2]) the effect of hole-hopping can be treated by including a larger set of modified exchange interactions in the neighborhood of each hole. Thus, the spin Hamiltonian of the doped system differs from that of the pure AF only in the strength of some exchange couplings:

$$\hat{H} = \hat{H}_0 - \lambda \hat{H}' \equiv \hat{H}_0 - \hat{V}, \quad (1)$$

where H_0 is the Hamiltonian for the perfect antiferromagnet, which, for concreteness, we take to have only nearest-neighbor interactions on a hypercubic lattice,

$$\hat{H}_0 = \sum_{\langle ij \rangle} \hat{H}_{ij}, \quad \hat{H}_{ij} = J \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \quad (2)$$

and the perturbation Hamiltonian \hat{H}' specifies a set of pairwise exchange interactions such that, in the limit $\lambda = 1$, a spin near which the hole is localized is disconnected from the rest of the system. Clearly, the *interaction* energy between hole clusters is obtained correctly in this limit, although the cluster self-energy could depend on the interactions between the fictitious, disconnected spins. We start with the simplest case of strongly localized holes, for which H' consists of the sum over the exchange interactions connecting the designated hole sites in a cluster to the nearest-neighbor sites surrounding it. One exceptional geometry which we treat differently is a stripe which is simultaneously an anti-phase domain wall in the AF order. Such a stripe can be treated [5], as shown in Fig. 2, effectively as a wall of *bonds* with altered exchange coupling, so that we work in the proper Hilbert space from the beginning; here naïve perturbation theory in powers of λ yields qualitatively correct generic results.

Casimir Energy of Stripes: We begin by considering the interaction energy between two stripes of static holes; this turns out to be the simplest problem because for $\lambda = 1$ the region between the two stripes is cut off from its surroundings, and, according to a very general argument originally due to Casimir [6], the interaction energy must fall off as $\mathcal{E} \sim -r^{-d}$. To be more explicit, if we take the stripes to be perfectly reflecting, the perpendicular component of the spin-wave's momentum in the region of width r between the stripes is discretely quantized, which modifies its vacuum energy. Then the distance-dependent part of the energy per unit hyperarea can be expressed as the difference

$$\mathcal{E}(r) = \sum_{k_\perp = \pi n/r} \int \frac{d^{d-1} k_\parallel}{(2\pi)^{d-1}} \left(\frac{\hbar\omega_{\vec{k}}}{2} \right) - r E_\infty,$$

where $\hbar\omega_{\vec{k}}$ is the spin-wave energy, and E_∞ is the vacuum energy density of the infinite lattice. The summation, performed with the Poisson formula, gives

$$\begin{aligned} \mathcal{E} &= r \sum_{m \neq 0} \int \frac{d^d k}{(2\pi)^d} \frac{\hbar\omega_{\vec{k}}}{2} e^{2ik_\perp r m} \\ &= -\frac{c}{r^d} \left[\frac{\zeta(d+1) \Gamma(d+1)}{2^{2d} \pi^{d/2} \Gamma(d/2)} \right] + \mathcal{O}(r^{-d-1}), \end{aligned} \quad (3)$$

which is proportional to the spin-wave velocity, c ($\hbar\omega_{\vec{k}} \approx c|\vec{k}|$; in linear spin-wave approximation [7] $c = JS/\sqrt{d}$), since only small momenta contribute to the large-distance asymptotics of the interaction energy. This result is universal: it depends on the number of acoustic modes and their speed but not on the form of the spectrum at high energies or the specific boundary conditions.

Linear Spin-Wave (LSW) Theory: In more general circumstances we need to compute the interactions from a more microscopic approach. For this purpose, we

adopt LSW theory which is quantitatively accurate for large S , but which we expect to be a reliable method for extracting the long distance physics for $d \geq 2$ even for $S = 1/2$, since already in $d = 2$ AF order is very robust [8]. In order to make contact with the perturbative results discussed below, we calculate the LSW correction to the ground-state energy in the presence of localized stripes or holes at arbitrary λ . The exact ground-state energy can be calculated as a coupling constant integral of the expectation value of the perturbation Hamiltonian using the Feynman-Hellman formula, or, in the LSW approximation, expressed as the functional determinant

$$-\mathcal{E}(\lambda) = \frac{1}{2} \text{Tr} \left(\ln \hat{\mathcal{G}}^\lambda - \ln \hat{\mathcal{G}}^0 \right). \quad (4)$$

The latter can be rewritten as a coupling-constant integral of the diagonal part of the exact LSW Green's function (GF) $\hat{\mathcal{G}}^\lambda$, as we shall see below.

LSW theory for stripes: As a first application of LSW theory, we recompute the interaction energy between two stripes oriented along the y axis in $d = 2$. In an obvious mixed representation, labelled by the conserved wave vector k_y parallel to the stripes and the lattice position x in the direction perpendicular to the stripes, the Dyson equation for the GF reduces to a finite sum,

$$\hat{\mathcal{G}}_{x,x'}^\lambda(\omega, k_y) = \hat{\mathcal{G}}_{x-x'}^0(\omega, k_y) + \lambda \sum_i \hat{\mathcal{G}}_{x-x_i}^0 \hat{\mathcal{G}}_{x_i,x'}^\lambda, \quad (5)$$

where i labels the vertical lines of sites connected by the weak bond representing the effect of the stripes, and the unperturbed GFs are given explicitly by

$$\hat{\mathcal{G}}_{\mathbf{r}}^0 = \begin{vmatrix} \mathcal{D}_{\mathbf{r}} & \mathcal{F}_{\mathbf{r}} \\ \bar{\mathcal{F}}_{\mathbf{r}} & \bar{\mathcal{D}}_{\mathbf{r}} \end{vmatrix} = \begin{vmatrix} \langle T_{\mathbf{r}} s_{\mathbf{r}}(\tau) \bar{s}_0(0) \rangle & \langle T_{\mathbf{r}} s_{\mathbf{r}}(\tau) s_0(0) \rangle \\ \langle T_{\mathbf{r}} \bar{s}_{\mathbf{r}}(\tau) \bar{s}_0(0) \rangle & \langle T_{\mathbf{r}} \bar{s}_{\mathbf{r}}(\tau) s_0(0) \rangle \end{vmatrix}, \quad (6)$$

where the operators $s_{\mathbf{r}} = b_{\mathbf{r}} + b_{\mathbf{r}+\hat{\mathbf{x}}}^\dagger$, $\bar{s}_{\mathbf{r}} = b_{\mathbf{r}}^\dagger + b_{\mathbf{r}+\hat{\mathbf{x}}}$ are defined on the bonds $\langle \mathbf{r}, \mathbf{r}+\hat{\mathbf{x}} \rangle$ in terms of the Holstein-Primakoff boson operators $b_{\mathbf{r}}$, $b_{\mathbf{r}}^\dagger$. As for the problem of a quantum particle in the presence of a finite number of point scatterers, the solution of Eq. (5) involves the inversion of only a finite matrix, and the functional determinant (4) can be rewritten in the convenient form,

$$\mathcal{E} = -\frac{JS}{2} \int_0^\lambda \frac{d\lambda'}{\lambda'} \int \frac{dk_y d\omega}{4\pi^2} \sum_i \left(\hat{\mathcal{G}}^{\lambda'} - \hat{\mathcal{G}}^0 \right)_{ii}, \quad (7)$$

with integrations over the coupling constant λ , the zero-temperature Matsubara frequency ω , and the conserved momentum k_y along the stripes, along with the finite summation over the stripe index i .

The matrix elements of the pure crystal GF (6) in the mixed representation are given explicitly by the integrals

$$\begin{aligned} \mathcal{D}_x(\omega, k_y) &= \int \frac{dk_x}{4\pi} \frac{1 - \gamma_k \cos k_x}{\omega^2 + \epsilon_k^2} e^{ik_x x}, \\ \mathcal{F}_x(\omega, k_y) &= \int \frac{dk_x}{4\pi} \frac{\cos k_x - \gamma_k - \omega \sin k_x}{\omega^2 + \epsilon_k^2} e^{ik_x x}, \\ \bar{\mathcal{D}}_x(\omega, k_y) &= \mathcal{D}_x(-\omega, k_y), \quad \bar{\mathcal{F}}_x(\omega, k_y) = \mathcal{F}_x(-\omega, k_y), \end{aligned} \quad (8)$$

where $\gamma_k = [\cos(k_x) + \cos(k_y)]/2$ and $\epsilon_k^2 = 1 - \gamma_k^2$. For the case of only two anti-phase stripes separated by the distance $r = |x_2 - x_1|$, the coupling-constant integration (7) can be performed analytically, with the result

$$\frac{\mathcal{E}}{JS} = \int \frac{d\omega dk_y}{8\pi^2} \sum_{\eta=\pm 1} \ln \frac{|\lambda^{-1} - \mathcal{D}_0 + \eta \mathcal{D}_r|^2 - |\mathcal{F}_0 - \eta \mathcal{F}_r|^2}{|\lambda^{-1} - \mathcal{D}_0|^2 - |\mathcal{F}_0|^2}. \quad (9)$$

The matrix elements of the bare spin-wave GF (6) are small at $r \rightarrow \infty$, so for all $\lambda < 1$, an asymptotic expression for the interaction energy can be obtained by expanding Eq. (9) in powers of \mathcal{F}_r , \mathcal{D}_r ; to leading order we obtain

$$\mathcal{E} = -\frac{JS\lambda^2}{r^4} \left[\frac{240\lambda(1-\lambda) + 3\sqrt{2}(51 - 102\lambda + 67\lambda^2)}{1024\pi(1-\lambda)^2[2 - (2-\sqrt{2})\lambda]^2} \right]. \quad (10)$$

The naïve perturbative results for this energy can be obtained by further expanding this expression to second order in λ :

$$\mathcal{E}^{\lambda \rightarrow 0}(r) = -\frac{JS\lambda^2}{r^4} \left[\frac{153}{2048\pi\sqrt{2}} \right] + \mathcal{O}(r^{-5}). \quad (11)$$

At $\lambda = 1$, however, the expression (10) diverges, because the denominator of the argument of the logarithm in Eq. (9) has a zero at the point $\omega = k_y = 0$. It is this zero, identified as the zero-energy spin wave state bound to each stripe, which is ultimately responsible for the modification of the asymptotic form of the interaction energy. As discussed later, the existence of such a state is related to the change in the structure of the Hilbert space as the clusters become isolated at this value of λ ; such a state exists near a hole cluster of any geometry, and it cannot be eliminated by corrections due to spin-wave interactions or to the holes' mobility. In our calculation, we account for this zero-energy state by solving the scattering problem near each stripe or hole cluster exactly.

The correct asymptotic behavior at $\lambda = 1$ can be obtained by re-evaluating the interaction energy, starting with the complete expression (9). Using the long-distance asymptotics of the components (8) of the GF, we obtain to leading order in $1/r$

$$\mathcal{E}^{\lambda=1}(r) = \frac{JS}{2r^2} \int \frac{dk_y d\omega}{(2\pi)^2} \ln \left[1 - e^{-2(k_y^2 + 2\omega^2)^{1/2}} \right],$$

which leads to the universal Casimir result (3) evaluated at $d=2$ and $c = JS/\sqrt{2}$. We have also investigated the crossover from the perturbative expressions (11), (10) at small values of λ to the universal form (3) at $\lambda = 1$ by integrating the exact LSW energy (9) numerically. The results are shown in Fig. 3, along with the corresponding asymptotic expressions.

LSW Theory for Isolated Holes: Zero-energy spin-wave bound states and the associated divergence of the perturbation series happen not only for extended objects

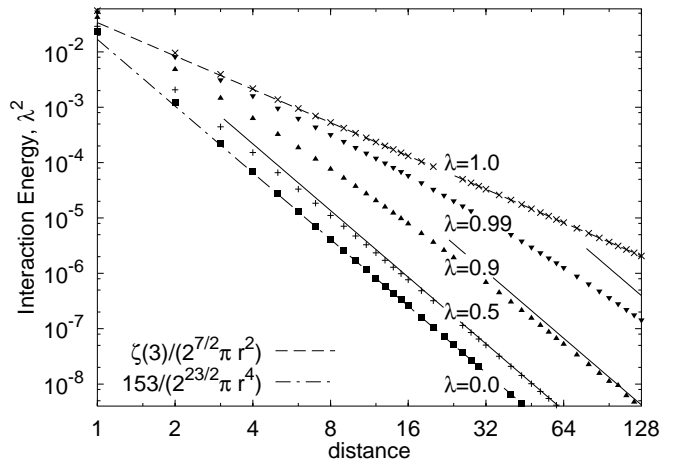


FIG. 3. The LSW Casimir energy of two stripes in units of $JS^2(\lambda^2/S)$ from Eq. (9). The $\lambda = 0$ line indicates the perturbative result. The solid lines show the calculated asymptotic behavior at $r \rightarrow \infty$ at intermediate values of λ .

like the stripes we just considered, but also for solitary holes or bigger hole clusters. As a second example, we consider explicitly the interactions between two isolated holes. The Hamiltonian of a single hole can be expressed as a sum of terms with different symmetries with respect to the point group [9], and we find that in arbitrary dimension, a zero-energy, totally symmetric bound-state emerges at the same critical value $\lambda = 1$, so that the corresponding spin-wave scattering amplitude diverges at small frequencies. Thus, while for any $\lambda < 1$, the interaction between two holes has the same asymptotic behavior as the leading order perturbative expression,

$$\mathcal{E}_{\text{pert}} = -\frac{JS\lambda^2}{r^{2d+1}} \left[\frac{(d^2-1)(d+1)\Gamma(d+\frac{1}{2})\Gamma(\frac{d+1}{2})}{2^{d-1}\pi^d d^{3/2}(d+2)\Gamma(\frac{d}{2})} \right], \quad (12)$$

for $\lambda = 1$ the asymptotic form of the interaction is

$$\mathcal{E}^{\lambda=1} = -\frac{JS}{r^{2d-1}} \left[\frac{\Gamma(d-\frac{1}{2})\Gamma(\frac{d+1}{2})}{2^d \pi^d \sqrt{d}\Gamma(\frac{d}{2})} \right], \quad (13)$$

which is valid both for holes at odd separations (different sublattices, holes with opposite spins) and holes at even separations (same sublattice, holes with the same spin).

Marginally-Bound States and the Proper Way to Do Perturbation Theory: We return now to the issue of the failure of naïve perturbation expansion in powers of λ . For a hole cluster of arbitrary geometry the Dyson equation [Eq. (5) for the case of stripes] gives bound states when

$$\det \left[\hat{\mathcal{G}}_{\mathbf{r}_i - \mathbf{r}_j}^0(\omega) - \delta_{ij} \lambda^{-1} \right] = 0, \quad (14)$$

where the two-component GF (6) is calculated for each pair of bonds present in the perturbation Hamiltonian \hat{H}' as specified in Eq. (1). At $\lambda = 1$, where the artificially introduced spins become disconnected, they acquire the freedom to rotate with respect to the rest of the system,

which reveals itself as a zero-energy spin-wave state localized on these spins. Because of the continuity of the GF in Eq. (14), this implies the existence of a soft spin-wave mode coupled to the holes, which is responsible for the divergence of the spin-wave scattering amplitudes off the hole cluster at small frequencies, and also provides the singularity required to modify the Casimir interaction energy obtained perturbatively.

We checked this argument by computing the eigenvalues of the matrix $\hat{1} - \hat{\mathcal{G}}_{\mathbf{r}_i - \mathbf{r}_j}^0 (\omega=0)$ for several hole cluster geometries, including those shown in Fig. 1. In agreement with the prediction, there are exactly two zero eigenvalues for the clusters with only one disconnected group of spins, one for each spin wave branch, and there are four such eigenvalues for the geometry in the second example in Fig. 1, where the two spins of the cluster are disconnected from each other. All other eigenvalues are positive, implying that the perturbation expansion is free of singularities for $\lambda < 1$. Within the same approach, we have also considered the effect of virtual hops of less strongly localized holes, which induce additional AF exchange couplings between the spins in the neighborhood of the hole. Once again, the zero-energy spin-wave bound state persists regardless of the strengths of these additional interactions.

Because the existence of a zero-energy spin wave bound state within an isolated cluster representing localized holes follows from symmetry arguments, it is a very robust feature of the spectrum. However, the degeneracy of the artificially inserted spins can be lifted by a local magnetic field term, added to the perturbation Hamiltonian \hat{H}' (obviously, this does not modify the physics of the rest of the system). It turns out that this term not only removes the divergence of the perturbation series, but also contributes an energy $\mathcal{E} \sim r^{-2d+1}$ to the interaction between the hole clusters already in second order of the perturbation expansion, which now gives the correct qualitative result. The improved perturbation expansion can be used to prove that the Casimir interaction between hole clusters indeed falls off as $\mathcal{E} \sim r^{-2d+1}$ even in the presence of spin-wave interactions.

The only case where the non-physical spins need not be introduced is the stripe serving as an anti-phase domain wall, modeled as shown in Fig. 2. Although perturbation theory here also breaks down at $\lambda = 1$ because of the soft spin-wave mode bound to the stripe, this mode is related to the freedom to rotate the AF magnetization of the two parts, which become disconnected at this value of λ . Such symmetry is easily destroyed by the holes' mobility or by spin-wave interaction corrections, and, therefore, the robust asymptotic form of the interaction energy between the antiphase stripes is the one given by the perturbation expansion (11). Note, however, that the general asymptotic behavior $\mathcal{E} \sim -J'Sr^{-d}$ is restored if next-nearest neighbor interactions are included in the Hamiltonian (2).

Implications: We have considered Casimir interactions between well separated hole clusters in AFs. For hole clusters or stripes in a uniform AF, this energy is uniformly attractive and generally falls off with distance as $\mathcal{E} \sim r^{-2d+1}$ and $\mathcal{E} \sim r^{-d}$ respectively. The interaction is quantitatively weak; for two holes in the $S = 1/2$ AF the interaction between next-nearest neighbor holes is less than $10^{-2}J$. However, because the interaction falls slowly with distance, it is important for an analysis of the stability of static charge-ordered structures in systems lacking long-range Coulomb repulsion. It has been conjectured [3,10] that phase separation is a ubiquitous feature of lightly doped antiferromagnets, and that consequently there is always a first-order transition separating the undoped and doped states. Evidence in support [11] and in conflict [4,12] with this conjecture has been obtained from numerical studies of small-size systems. Phase separation has been shown to occur [13,14] in the large d limit of the Hubbard and t - J models, and in the mean-field spiral states of the large N t - J model [15]. The present results offer strong additional support for the validity of this conjecture. Specifically, we claim that because of this Casimir-like interaction, any static ordered state of neutral holes will be thermodynamically unstable with respect to phase separation at small enough doping.

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